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Experimental Design for Reflection
Measurements of Highly Reactive Liquid
or Solid Substances with Application to
Liquid Sodium

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ABSTRACT

This technical report describes the experimental part of program on thermal radiation properties of reactor materials. The report is based on the M.S. Dissertation submitted by A.A. Gossler, Department of Mechanical Engineering, University of Wisconsin at Milwaukee, June 1980.

A versatile goniometer system with associated electronic components and mechanical instruments has been assembled. It is designed to measure spectral, specular reflectances of highly reactive liquid or solid substances over a spectral range of 0.3μ to 9μ and incidence angles of 12° to 30° off the normal direction. The capability of measuring reflectances of liquid substances clearly distinguishes this experimental design from conventional systems which are applicable only to solid substances. This design has been used to measure the spectral, specular reflectance of liquid sodium and preliminary results obtained are compared with those of solid sodium measured by other investigators.

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NOMENCLATURE

A	Reflected Beam in Figure 7
B	Reflected Beam in Figure 7
D	Length Defined in Figure 7
E	Total Possible Error
H	Length Defined in Figure 7
k	Complex Portion of Refractive Index
IR	Infrared Wavelengths
L	Length Defined in Figure 7
LIA	Lock-In Amplifier
LED-PD	Light Emitting Diode Phase Detector
n	Real Portion of Refractive Index
$\hat{n}$	Complex Refractive Index: $\hat{n} = n + ik$
S	Signal
T	Length Defined in Figure 7
UV	Ultra-Violet Wavelengths

Subscripts

i	Incident
int	Internal
o	External
r	Reference
s	Sample
t	Transmitted
x	Reference or Sample

Greek Symbols

κ	A Constant
λ	Wavelength
ρ	Reflectance
τ	Transmittance
θ	Angle

I. INTRODUCTION

Temperatures involved in nuclear reactor safety analysis are in the range of 1300° to $10,500^{\circ}$ F indicating that radiative heat transfer may be a dominant heat transfer mode. Essential to radiative heat transfer calculations is accurate information on the thermal radiation properties of the associated materials. Particularly, to calculate surface radiation properties, the optical constants of a material's complex refractive index must be known. A work done by Didrikil¹ developed a method of analysis for determining these optical constants from spectral reflectance data at measured incidence angles for non-polarized light. This method simultaneously eliminates the need for a polarizer and increases the intensity of the reflected light, thereby overall simplifying the required experimental equipment.

Fast breeder nuclear reactors propose to use liquid sodium as the primary loop working fluid. While a large amount of work has been done beginning with P. Drude circa 1890, as cited in ref. 2, to determine the optical constants of almost all metals, sodium is one of the few exceptions. Due to its rapid oxidation, the production and preservation of a plane, bright metallic surface is connected with extraordinary

experimental problems.

Efforts to date have established optical constants for solid sodium only at selected wavelengths.^{2,3,4} Very recently, Chan and Tseng⁵ made use of the optical constants of solid sodium and calculated analytically the reflectance between sodium and other reactor materials. It is apparent that no direct measurement of radiation properties of liquified sodium has yet been made, though the reactor safety analysis deals with liquid sodium. The primary purpose of this study is to design a unique goniometer system capable of measuring the spectral, specular reflectance of a liquid substance such as liquid sodium. Preliminary results obtained are discussed in comparison with the information on solid sodium obtained to date by other experimenters.

II. EXPERIMENTAL SET-UP

The experimental design for measuring optical properties is divided into several subsystems. A classification scheme based on functional distinctions of the design's major elements readily lends itself to a detailed description of the overall system. Hence, this paper describes in practical sequence each subsystem and its interface with other subsystems. It is intended that this will lead to a clear understanding of the system as well as a useable guide for its future use.

II.1 Double-Beam Goniometer

The experimental arrangement employs a vertically mounted double-beam goniometer, (see Figure 1), which houses the signal source. Along with the signal source the goniometer consists of a signal source 'chopper', a beam divider, and guiding and focusing optics. Its purpose in general terms is to provide a broad spectrum electromagnetic or 'light' signal, channel this signal appropriately and ultimately refocus the signal and direct it to the monochromator. This subsystem is variable in a range of 12° to 30° angle of incidence to the sample being investigated, 0° to 150° circumferential angle, and allows optical measurements of

specular reflection over these ranges or measurements of transmittance over a given wavelength range.

II.1.1.1 Light Source

The light source, known as a Nernst glower, is a tungsten ribbon in a quartz envelope which emits electromagnetic energy from the ultra-violet (UV) to the far infrared (IR) when a current is passed through it. It was selected for its small size, approximately 14mm by 1½mm, which allows sharp focus, and for its high intensity output at low power input. Output intensity is easily adjusted by use of a connected unit called a Nernst glower supply. This is a simple circuit design in which the current through the glower is step-wise increased by switching on successive common household light bulbs, termed ballast bulbs in this application. These bulbs are in series with the glower and in parallel with each other. With 25 watt bulbs in the circuit, each of the 3 switchable current regulating ballast bulbs increases the current through the glower bar by approximately 0.2 amperes. With 40 watt bulbs replacing the 25 watt bulbs an increase of slightly greater than 0.3 amperes per bulb results, with a total possible current of approximately 1.0 amperes--the maximum allowable current and so the maximum allowable intensity without significant

decrease in the glower life. To turn the glower on, a momentary switch is depressed which engages small preheater coils located on either side of the glower as the maximum current is passed through it. In approximately 20 seconds the glower bar indeed begins to glow and the preheater is disengaged.

II.1.2 Signal Modulation and Direction

The source signal emitted by the glower is immediately 'chopped' by a rotating cut metal disk which regularly blocks the signal entirely. This disk is driven by an electric motor synchronous with line frequency and modulates the signal to 102.9 complete on-off cycles per second. This disk is located approximately one centimeter in front of the source housing. It is placed before the sample in order not to chop radiation emitted by a heated sample. That is, the signal intensity at this point is indeed solely a function of the source strength. A light emitting diode phase detector (LED-PD) pick-up detects the signal modulation for use as a lock-in amplifier (LIA) reference signal. In this way the LIA isolates the source intensity and instantly monitors any fluctuations in the source intensity independent of ambient conditions. This is described in greater detail in the Signal Processing section.

The chopped signal is then directed and focused by one adjustably tilting flat mirror and one adjustably tilting and telescoping parabolic mirror and strikes the beam divider. These and all other mirrors in the goniometer subsystem are first surface evaporated aluminum mirrors the reflectance of which is spectrally known³ and assumed here to be specularly constant over the range of incidence angles physically obtainable by the goniometer. A first surface mirror here denotes a mirror with the reflective surface exposed to incident light as opposed to a conventional mirror which protects the reflective surface by a layer of glass.

It is the beam divider which defines the goniometer as a double-beam goniometer. The beam divider is also commonly referred to as a butterfly mirror. It is a cut glass disk with one mirrored surface; it rotates rapidly alternating the chopped light source between the sample channel and the reference channel at the rate of 17.1 Hz. This eliminates the effects of atmospheric absorption bands as the light source travels the same optical length in each channel. The beam divider's rotation is governed by a line synchronous motor and is detected by another LED-PD pick-up for use as a reference signal in a second LIA. This allows instant monitoring of the difference in signal

intensities between the sample and the reference channels. Again, details of this are included in the Signal Processing section.

The sample channel contains the sample being investigated. The reference channel utilizes a reference mirror, a first surface evaporated aluminum mirror as mentioned previously. The goniometer allows for these two channels to be physically interchanged. One channel, the upper channel, that is, the channel physically above the beam divider, necessarily has 2 mirror reflections associated with it attributed to the beam divider's mirrored surfaces. The reference mirror also has 1 mirror reflection associated with it. In the measurement of the reflectance of liquid sodium, the upper channel was used as the reference channel and therefore the reference signal leaving the beam divider was equal to the source signal multiplied by a factor of a known reflectance raised to the third power.

Leaving the beam divider the signal is refocused by another adjustably tilting and telescoping parabolic mirror and directed by an adjustable flat mirror to a set of three mirrors known as a signal rotator. This properly orients the signal to the variable width slit opening of the monochromator. With a total of

10 mirrored surfaces, dust accumulation is a potential problem. In order to maintain the maximum signal strength and true readings the mirrors should be virtually dust-free. However, being first surface, polishing or even wiping of the surfaces significantly and permanently damages the reflective quality of the mirrors. For this purpose a collodion solution used to clean telescope mirrors was used with great success. The solution quickly dries trapping particulates and is peeled off taking the particulates with it without affecting the delicate mirror surface.

II.2 Monochromator

The purpose of the monochromator is analogous to that of a radio tuner and is to isolate a narrow bandwidth of electromagnetic energy about a known wavelength and to focus this on a signal detector housed in the monochromator. The entering signal passes through a slit continuously adjustable from 5 to 2000 μ with height limitation from 0 to 20 mm. It is then reflected off a stationary collimating first surface mirror onto a reflection grating at a very precise incidence angle. The wavelength of the energy reflected off this grating onto a stationary focusing first surface mirror is a function solely of the incidence angle for a given grating. The spacing

and blazing of the grooves on the grating determines the wavelength of the reflected signal at a given incidence angle. Generally, several spectra are so produced causing spectra overlap and reduced intensity because the available energy is divided among a number of spectra. However, a large fraction of the energy can be concentrated in a particular spectrum or 'order'; this is known as blazing. The monochromator made use of interchangeable grating of different groove spacings and blazings ideally resulting in a continuous spectrum from 0.3 to 9.0 μ . However, in the course of time one grating surface was apparently damaged and UV readings from 0.3 to 1.0 μ could not be taken. This underscores the need for careful handling of the gratings. Mechanical contact with a solid, liquid, or vapor damages the grating. Hence, even breathing or talking over the surface is ruled out.

Gratings of the same line spacing can be interchanged with no adjustments necessary. When a grating of different line spacing is installed, the gears on the drive and screw shafts located at the rear of the instrument must be changed according to manual instructions. In this way the wavelength counter and speed control settings continue to read correctly. The angle of incidence to the grating is controlled by

an electric 12 speed reversible synchronous motor housed in the monochromator which simultaneously controls the mechanical counter also housed in the monochromator. The counter displays the wavelength being investigated. The motor allows scan rates from 0.5 to 2000 Å/minute with a 5 digit wavelength read-out.

A focusing stationary mirror finally directs the signal to a completely adjustable telescoping parabolic first surface mirror which in turn focuses the signal onto the signal detector.

II.3 Signal Processing

Generally, the purpose of the signal processing subsystem is to yield and record a ratio of the unknown but measured optical property of the sample to a known quantity. In measuring the reflectance of liquid sodium the desired quantity was the ratio of the liquid sodium reflectance to the reference mirror reflectance cubed. This subsystem is also shown on Figure 1.

II.3.1 Signal Detector

The signal detector, a transducer, is a thermopile with a ZnSe window. It was selected

for its overall sensitivity to wavelengths from the UV to the far IR and for its short response time. The detector has a low impedance and a very low current tolerance. Attempting to measure the impedance of the detector with a common ohm-meter results in the detector's destruction. Also, careless handling of the detector can cause its destruction as a result of static charges. For the safe handling of the detector it is necessary to wipe all surfaces including hands brought into contact with it with alcohol first to eliminate static charges. Plastic surfaces also need to be avoided as they easily generate electrostatic charges. These precautions were not followed on one occasion and the thermocouple was in fact damaged. This sort of damage, which incidentally does not fall under the company's warranty policy, required return to and repair by the vendor with all the related paperwork and delay. Careful handling of this detector is strongly encouraged.

This signal detector converts the optical signal to an electrical one and transmits it to an impedance matching transformer.

II.3.2 Transformer

The transformer which interfaces both the

signal detector and a preamplifier has voltage gains of x1000 and x100 and as mentioned its primary purpose is to match the low output impedance of the signal detector with the optimum input impedance of the preamplifier. Running an arbitrary impedance source directly into an amplifier or preamplifier may result in excessive noise. By using the x1000 gain on the transformer, the low impedance signal detector is brought into the minimum noise range of the preamplifier.

II.3.3 Preamplifier

The preamplifier, a nanovolt preamplifier, results in an additional x1000 voltage gain. The output of the preamplifier is fed into 2 parallel LIAs. An argument can be made that the use of the preamplifier may be superfluous in most instances and so should be removed from the signal processing system. That is, the gain introduced by the preamplifier is generally not needed while complicating a high gain circuit generally introduces additional noise. However, it is found that the noise introduced by the preamplifier after amplification by the LIAs is several microvolts and the signal being measured (such as in the liquid sodium experiment) is in the millivolt or greater range. The noise of the preamplifier is insignificant being at least 3 orders of magnitude smaller than the signal

being measured. Therefore, though the preamplifier gain is needed only intermittently if at all, it is practical to leave it in the system as opposed to removing it and replacing it when called for.

II.3.4 Lock-In Amplifiers

The function of a LIA is to isolate and amplify a signal at a given frequency. The amplifier 'locks in' to a given frequency known as the reference frequency, amplifies only the rms value of the signal varying at the reference frequency and outputs a dc signal proportional to this value. In the liquid sodium experiment two such amplifiers were used and will here be denoted by LIA-1 and LIA-2.

As was mentioned LED-PD pick-ups at the source chopper and at the beam divider detect the frequency of the signal modulation at these points. These pick-ups are connected to a controlling electronic unit which engages the pick-ups and the synchronous motors which drive the source chopper and beam divider. (Refer to Figure 1.) This unit also serves to amplify the LED-PD pick-ups' signals. The controlling unit can be treated as a fixed operational amplifier. When the beam emitted by the pick-ups is interrupted a zero dc reference voltage is output. When the

beam is detected by the pick-ups a dc voltage of about 4.8 volts is output. The result is two periodic functions about 4.8 volts peak to peak at the source chopper or beam divider frequencies that are roughly square in shape. This unit is specifically designed for use with LIAs and the produced signal is sufficient to trigger any common LIA reference input.

LIAs function by inverting the input signal at the reference signal frequency, as shown in Figure 2. The output is the amplified rms value of the modified input signal. An adjustable time constant smooths out the output to a constant dc level. It is clear that the phase relationship between the reference signal and the input signal is significant. This relationship is continuously adjustable by a front panel control knob on the LIA.

In the liquid sodium experiment the reference frequency of LIA-1 was that of the signal modulation rate due to the source chopper. Figure 3 schematically depicts these circumstances. It is assumed in this figure that the reflectance of the reference mirror cubed is higher than that of the sample, hence $S_r > S_s$. It is seen that the output of LIA-1 was proportional to $\overline{S_r} + \overline{S_s}$.

The second lock-in amplifier, LIA-2, was referenced to the beam divider and so amplified only the rms value of the signal varying at the reference frequency. Figure 4 represents this. The output of LIA-2 was proportional to $\overline{S_r} - \overline{S_s}$.

II.3.5 LIA Operation

The LIAs are operated as follows. When a signal is detected an appropriate full-scale sensitivity and time constant are selected. The shortest time constant which yields a stable output signal should be used. The phase between the reference and the input signals is adjusted until the output reading goes to zero. At this point the reference and input signals are 90° out of phase. The offset should then be checked. A front panel button changes the reference signal and input signal phase relationship by 180°. If after depressing this button the output reading is not still zero the offset is adjusted so that any reading fluctuates symmetrically about zero as the reference to input signal phase relationship is repeatedly changed by depressing this 180° phase relationship button. When the offset has been adjusted the phase relationship is again adjusted by the continuously adjustable control knob so that the output is zero. Changing the phase relationship by 180° at this time

should not affect the zero output. This phase relationship is now changed by 90° by another front panel button. This gives the maximum output reading. The phase relationship can now be changed by 180° resulting only in a sign change of the output. The full scale output of the LIAs is ± 1 volt.

II.3.6 Auxiliary Amplifier; Equalizing LIAs

The outputs of LIA-1 and LIA-2 are input to an auxiliary amplifier unit. Due to the response time constants of the signal detector and transformer and to the different wave shape (i.e. rms values) resulting from the different signal modulators and to the frequency dependent output of LIAs, the signal output from the LIAs must routinely be adjusted. That is, by blocking the signal in the goniometer sample channel, the equivalent of using a zero reflectance sample, the LIA outputs should be equal. The schematic labelled Figure 5 indicates this. However, due to the reasons cited above, this is not the case and the auxiliary amplifier unit adjusts for this. In fact, exactly this procedure was followed to equalize the LIA outputs for the liquid sodium measurements.

The auxiliary amplifier is a variable arithmetic analogue unit with separate gain controls for each of

the two inputs. The unit amplifies, or 'adjusts', the inputs and yields three separate output signals. Two of the output signals are the negative values of the amplified inputs: With inputs proportional to $(S_r + S_s)$ and $(S_r - S_s)$, the adjusted outputs are some constant κ multiplied by $-(S_r + S_s)$ and $-(S_r - S_s)$, respectively. The third output is the sum of the amplified inputs: The inputs of $(S_r + S_s)$ and $(S_r - S_s)$ yield an output of $\kappa\{(S_r + S_s) + (S_r - S_s)\}$.

II.3.7 Ratiometer

The output signals of the auxiliary amplifier are then processed by a ratiometer which is another fixed arithmetic analogue unit. The ratiometer has three inputs and yields a ratio of the difference between 2 input values divided by the third input value. With inputs of $-\kappa(S_r + S_s)$, $-\kappa(S_r - S_s)$, and $\kappa\{(S_r + S_s) + (S_r - S_s)\}$ this unit has as its output

$$\frac{-\kappa(S_r + S_s) - \{-\kappa(S_r - S_s)\}}{\kappa\{(S_r + S_s) + (S_r - S_s)\}} = \frac{S_s}{S_r}$$

This ratio is simply that of the sample channel signal strength to the reference channel signal strength.

II.3.8 Recorder

The output of the ratiometer is recorded on a

strip chart recorder. Hence, in the liquid sodium application, the strip chart output at any point multiplied by the known reference mirror reflectance cubed yielded the liquid sodium reflectance.

II.4 Sample Holder Vacuum Subsystem

In the case of liquid sodium a 5 gram sample had to be carefully handled. Heated to approximately 225° F the metal was spontaneously and explosively combustible in air, reacting with the water vapor and oxygen in the air. A CO₂ fire extinguisher was kept at hand. The sample holder subsystem was designed to safely maintain pure liquid sodium.

To accomplish its purpose the sample holder vacuum subsystem held the liquid sodium, initially 99.95% pure in a vacuum of 1×10^{-6} torr. (One torr equals 1 mm of mercury.) Figure 6 is a schematic of this subsystem.

II.4.1 Sample Holder Vacuum Subsystem Operation

The operation of this subsystem is as follows. The sample holder section is removed from the system by closing valve #1. Valves #2 and #3 are opened to the system. It is important to note that valve #3 can be opened to the atmosphere or to the system.

If valve #3 is opened to the atmosphere at this point the mercury will be forcefully pushed into the delicate MacCleod gage when the vacuum is initiated. This will undoubtedly shatter the glass gage. The cooling jackets are then turned on slowly. The first cooling jacket condenses oil vapors escaping from the mechanical pump and mercury vapor diffusing away from the diffusion pump. By trapping the mercury vapors here the mechanical pump oil is kept clean assuring optimum performance as well as minimizing the possibility of mercury vapors getting into the atmosphere. The second cooling jacket condenses mercury in the diffusion pump for re-use.

The mechanical pump should then be engaged. The operating manual instructions should be checked to be sure the proper level of clean oil is in the pump. The oil level in this pump is routinely higher when operating than when turned off. The ultimate vacuum produced by this pump is published to be 10^{-4} torr. In actual operation approximately this vacuum is found to be maintained by this pump.

Liquid nitrogen is then poured into the cold-trap Dewar. Liquid nitrogen at -321° F serves to condense mercury vapors escaping from the diffusion pump or MacCleod gage when in use and other condensable gases

in the system (e.g. water vapor, oxygen). Because of these condensed vapors it is imperative that the system be vented when the vacuum pumps are turned off and the nitrogen allowed to boil off. Otherwise, rapid expansion of these condensed gases can result in an explosion.

The diffusion pump is then engaged by heating the mercury reservoir to its operating temperature. It was found the optimum performance was obtained by establishing 100 volts across the permanently installed heating element. About 25 minutes are required until the diffusion pump is operating at capacity at which time mercury will be seen condensing in cooling jacket #2. If a 10^{-6} torr vacuum is not obtained in 24 hours a leak is present in the system. In this case joints and valves should first be systematically inspected followed by a search for hairline cracks or pinpoint holes. For this purpose a high voltage gas ionizer or 'Tesla coil' should be used. This device ionizes rarefied gases which then give off characteristically colored light. The 24 hour 'pump down' period is necessary to completely de-gas the inner surfaces of the vacuum system.

At this point (and at any succeeding point in time) the MacCleod gage is brought into the system in

order to measure the vacuum achieved. To do this valves #2 and #3 are closed. Valve #2 is closed to prevent mercury from accidentally climbing completely through the gage and into the rest of the system. Valve #3 is then slowly opened to atmosphere. If quickly opened the MacCleod gage will be destroyed in the manner previously described.

There are three scales on the MacCleod gage corresponding to three ranges of vacuums. The gage has on it the proper mode of levelling and reading the gage. The mercury is held at a given level by closing valve #3. Basically, the gage uses atmospheric pressure to compress an exact volume of rarefied gas. The compression obtained as measured by a precise column of mercury indicates the vacuum.

After the measurement is taken valves #2 and #3 are opened simultaneously to the vacuum system. Both valves are opened to equalize the pressure in the gage. If this is not followed and the pressure in the system changes drastically or has changed drastically during the measurement (e.g. system failure) the mercury will either be forced into the rest of the system or will 'blown down' into the glass reservoir, shattering it. This blown down phenomenon occurred once during the testing of the system. The mercury reservoir

shattered, spewing mercury over a considerable area as well. Close attention to the operation of the MacCleod gage is clearly called for. The MacCleod gage should always be taken out of the system by closing valves #2 and #3 when not being used. Otherwise, mercury vapor will permeate the system and reduce the ultimate vacuum attainable. (The vapor pressure of mercury⁶ at room temperature is approximately 10^{-3} torr.)

The section beyond valve #1 can be called the sample holder proper and is discussed at this point. The sample holder proper consists of the sodium ampule, the testing cell and heating components.

The testing cell has a KBr window chosen for its transparency from the IR to the UV attached by a slow drying epoxy which maintains its integrity during heating under vacuum. Before being epoxied, the window which is highly hygroscopic must be polished to remove a thin cloudy water layer which accumulates to some degree due to the water vapor in the air. This polishing is accomplished by light rubbing on a felt or velour material wetted with a small amount of alcohol. Once secured in place the window's inner surface will not fog up once the vacuum is established. If the outer surface fogs up it can be polished carefully while in place. Also, if light marks or

fingerprints are accidentally deposited on the window surface at this point in time the same procedure can be used to remove these marks. To maintain equal optical conditions in the sample and reference goniometer channels a KBr window of the same thickness was introduced in front of the reference mirror.

II.4.2 Handling the Sodium Ampule

The sodium ampule consists of a small glass capsule with a break-seal inside a larger long glass tube. After the initial 24 hour pump down the vacuum must be checked to be certain 10^{-6} torr has been achieved. Proceeding with a vacuum of somewhat lower quality (e.g. 5×10^{-5} torr) results in a slow reaction at the surface of the liquid sodium making it useless in a matter of one to two hours and possibly much sooner as experience has shown. (See Results and Discussion section.) When the vacuum is found to indeed be 10^{-6} torr the ampule is removed from the system at its joint, epoxy is applied at the joint, and the break-seal is broken with a long slender instrument. The ampule is immediately replaced and valve #1 is quickly opened to the system. Again, a 24 hour period must be observed to allow for de-gassing. The vacuum must again re-establish at 10^{-6} torr or there is no point in proceeding. The vapor pressure of

sodium⁶ at 215° F is 1.5×10^{-7} so that introduction of the sample does not decrease the ultimate vacuum obtainable. If the system has no leaks and the total of 48 hours has been observed the system should be ready for liquification of the sodium.

II.4.3 Liquifying the Sodium

To liquify the sodium the testing cell which is controlled by one heating element and the ampule section which is controlled by another heating element are each heated by slowly increasing the voltage across the elements with a variac. All heating must be done very slowly to avoid cracking of the glass. Temperature in the experiment was monitored with iron-constantan termocouples and a potentiometer. The heating process took approximately one hour. The melting temperature of sodium is about 215° and melting temperatures of its oxide or hydroxide are several hundred Fahrenheit degrees higher. Thus only liquid sodium drained into the testing cell. The heating coil about the sodium ampule section was then turned off and only the cup remained heated.

II.5 KBr Window Effects

The possible effect of the KBr window on the experiment results invites scrutiny. The real portion

of the index of refraction of KBr^{3,4} in the range of 0.4 to 25 μ varies smoothly from 1.59 to 1.46 and is virtually invariant with temperature from 70 to 250° F. Using this information with Snell's law and Fresnel's formulae, the reflectivity of KBr is determined⁷:

$$\hat{n}_{\text{air}} \sin \theta_i = \hat{n}_{\text{KBr}} \sin \theta_t$$

where subscripts i and t refer to incident and transmitted, respectively. Using representative values of $\theta_i = 15^\circ$, $\hat{n}_{\text{KBr}} = 1.52 + 0i$ the transmitted angle is

$$\theta_t = 9.8^\circ$$

The internal reflection of the KBr window can now be found, according to Fresnel⁷:

$$\rho_{\text{int}} = \frac{1}{2} \left[\left(\frac{\hat{n}_{\text{KBr}} \cos \theta_i - \hat{n}_{\text{air}} \cos \theta_t}{\hat{n}_{\text{KBr}} \cos \theta_i + \hat{n}_{\text{air}} \cos \theta_t} \right)^2 + \left(\frac{\hat{n}_{\text{air}} \cos \theta_i - \hat{n}_{\text{KBr}} \cos \theta_t}{\hat{n}_{\text{air}} \cos \theta_i + \hat{n}_{\text{KBr}} \cos \theta_t} \right)^2 \right]$$

$$\rho_{\text{int}} = 0.043$$

Figure 7 pictures a light beam penetrating the KBr window reflecting off a surface, represented by a thick horizontal line, and again penetrating the window. This beam of light represents that reflecting off the reference mirror or that reflecting off a sample of

interest and the subscript x refers to this condition. This beam of light (drawn with an arrow in Figure 7) will here be referred to as the desired beam as it is this beam which yields desired rather than erroneous information. Also shown is the external window reflection, labelled 'A' and referred to by the subscript 'o', and the first, second and tertiary internal reflections. It is evident in the figure that if internal reflection B does not coincide with the desired beam then the external reflection A also will not coincide with the desired beam. The actual light beams schematically represented have finite widths. The Nernst glower provides a beam which is focused in the double-beam goniometer at the reference mirror or sample surface to a width of approximately $1700\ \mu$. This image is ultimately focused onto the variable width slit opening of the monochromator. The slit width is adjusted to be slightly narrower than the beam width. Hence, if distance D labelled on the figure is $1700\ \mu$ or greater this assures that the first internal reflection B and the external window reflection A will not falsely enter the signal detection and processing subsystem. Therefore, the largest possible single erroneous signal intensity due to window reflections

internal or external would be no larger than 0.043^2 or 0.18% of the incident signal. Distance D at a given angle is a function of height H as seen in Figure 7. All notation below refers to the same figure.

$$L = 2(H \tan \theta_i) \quad (L \neq L(\theta_t, T))$$

$$D = L \cos \theta_i$$

$$D = 2H \tan \theta_i \cos \theta_i$$

$$D = 2H \sin \theta_i$$

Therefore,
$$H = \frac{D}{2 \sin \theta_i} .$$

If $\theta_i = 15^\circ$ and taking D as 1700 μ

$$H = \frac{1700 \mu}{2 \sin 15^\circ} = 3284 \mu \approx 3.3 \text{ mm.}$$

Hence, as long as a minimum distance of 3.3 mm is maintained between the sample and the KBr window or between the reference mirror and the KBr window, reflections due to the window are insignificant. This distance was kept at a minimum of 18 mm in the liquid sodium experiment. Appendix A derives a closed form solution for the total possible error introduced by the window's multiple reflections and this is found to be 0.00136.

III. SYSTEM CALIBRATION

As a test of the accuracy of the system the reflectance of an evaporated aluminum surface was experimentally determined. By inserting such surfaces in both the sample and the reference channels the ratiometer yielded and the strip chart recorded the reflectance of the surfaces squared because of the two such reflections associated with the beam divider (as described in the Double-Beam Goniometer section). Table 1 shows these results and the results obtained by G. Haas, as cited in ref. 3, for the reflectance of a freshly evaporated film of evaporated aluminum. The experimentally determined results show very good agreement with results being very repeatable. The results obtained using the system described in this work were zero to $1\frac{1}{2}$ per cent less than those given by Haas. This is likely due to actual differences in the surfaces measured. Primarily, the surfaces measured in this experiment were not 'fresh'.

IV. RESULTS AND DISCUSSION

The reflectance of liquid sodium as measured to date by this double-beam goniometer system is presented in Table 2. More extensive measurements were not possible at this time due to a small crack discovered in the mercury reservoir of the MacCleod gage. This vacuum leak apparently developed and was found shortly after the beginning of the measurements and it appeared to become progressively worse as time progressed. The run was aborted because the liquid sodium sample was oxidizing at its surface and also because a catastrophic vacuum system failure was a possibility.

Due to sodium's large surface tension and low density the sample formed an ellipsoid shape about one inch in width. A 2 mm by 1.6 mm rectangular surface area at the center of the ellipsoid was used for the measurements. This appeared to approximate a flat surface well.

The sodium sample looked pure as it first entered the test cell. The measurements were begun at 1.5 μ wavelength and proceeded towards the longer IR wavelengths. The measurements from 1.5 to 3.0 μ were completed and immediately repeated. The remaining measurements were then completed. Hence, due to

gradual oxidation of the sample, the first set of measured values is the most reliable. The reflectance of the liquid sodium so determined is presented in Table 3. Measurements taken later in the experiment (beyond 3.0μ) are questionable as distinctive gray spots of sodium oxide were evident at the sample surface. In an attempt to clear the surface the liquid sample was agitated on several occasions. This met with no success. This may, however, account for the scatter in the measurements in this range as the agitation did move the oxide spots about the surface.

Althoff and Hertz⁸ present experimental values of the complex refractive index for solid sodium. Using these value in Fresnel's formulae yield the reflectance of the sodium. This information is presented in Table 4.

It is unlikely that the discrepancies between Tables 3 and 4 can be attributed solely to the physical state of the liuquid sodium. Even though the liquid sodium sample in this experiment seemed pure with a bright metallic surface for the measurements presented in Table 3, it is possible if not likely that due to the detected leak a thin oxide layer existed on the surface. The possibility exists, too, that the flat surface approximation is not accurate. That is,

taking the roughly 2 mm square surface area at the center of the ellipsoid shaped liquid sample may incorporate significant error. Decreasing this area by using a smaller slit width and height at the monochromater entrance, hence, approximating a point reflection more closely, may require a stronger light source.

V. RECOMMENDATIONS FOR FURTHER WORK

Further measurements are recommended upon replacement of the mercury reservoir. These measurements can be completed over several incidence angles. The sodium should then be allowed to solidify at room temperature and measurements could then be repeated. In this manner the effect of the phase change should be immediately evident and comparison of these data with existing data will be direct. It is only after these measurement that determination of the optical constants of liquid sodium can proceed with confidence.

Table 1

Measured and Previously Published Evaporated Aluminum
Reflectance Values

λ, μ	Published Values (ref. 3) ρ_{Al}	Measured Ratio $\rho_{Al}^3 / \rho_{Al, \theta_i = 15^\circ}$	ρ_{Al} (Average)
1.0	93.9%	.880, .874, .865	93.4%
1.5	96.8	.904, .904, .903, .910	95.2
2.0	97.2	.908, .908, .903, .903	95.2
3.0	97.5	.922, .922, .922	96.0
4.0	97.6	.923, .925	96.2
5.0	97.7	.937	96.8
6.0	97.7	.924	96.1
7.0	97.8	.935	96.7
8.0	97.9	.952	97.6

Table 2

Preliminary Results; Liquid Sodium Reflectance at 15° Incidence

λ, μ	Measured Ratio $\rho_{\text{Na}} / \rho_{\text{Al}}^3$	ρ_{Na} (Average)
1.5	.620, .544	50.2%
2.0	.600, .502	47.5
2.5	.570, .487	46.2
3.0	.550, .452, .455	43.0
3.5	.440	42.3
4.0		
4.5	.373, .380	33.7
5.0	.398, .384	30.1
5.5	.395, .389	31.6
6.0	.382, .440	36.5
6.5	.422, .500	41.3
7.0	.370, .503	39.5
7.5	.330, .470	36.7

Table 3

Preliminary Results; Initial Measurements of Liquid
Sodium Reflectance at 15° Incidence

λ, μ	Measured Ratio $\rho_{\text{Na}} / \rho_{\text{Al}}^3$	ρ_{Na}
1.5	.620	53.5%
2.0	.600	51.8
2.5	.570	49.8
3.0	.550	48.7

Table 4

Reflectance of Solid Sodium at 15° Incidence Based
on Althoff and Hertz.

λ, μ	ρ_{Na}
2.5	98.7%
3.0	98.9
3.5	98.9
4.0	98.9
7.2	98.6

Figure 1 Double-Beam Goniometer and Signal Processing Subsystems.

Figure 2 Relationship of LIA Input Signal and Reference Signal.

Figure 3 Representation of the LIA-1 Modulated Signal With a Non-zero Reflectance Sample.

Bars Denote rms Values

Figure 4 Representation of the LIA-2 Modulated Signal With a Non-zero Reflectance Sample

Bars Denote rms Values

Figure 5 Representation of the Modulated Signal With a Zero Reflectance Sample.

Figure 6 Schematic Representation of the Sample Holder Vacuum Subsystem.

Figure 7 Signal Reflections and Transmissions Associated With the KBr Window.

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APPENDIX A -- Total KBr Window Effects

The multiple reflections occurring at the KBr window are shown in Figure 7. Summing all reflections gives the following expression:

$$\begin{aligned} & \rho_o + \rho_{int}(1-\rho_o)(1-\rho_{int})\tau^2(1+\rho_{int}^2\tau^2+\rho_{int}^4\tau^4+\dots) \\ & + \rho_x(1-\rho_o)^2(1-\rho_{int})^2\tau^2(2\rho_{int}^2\tau^2+3\rho_{int}^4\tau^4+\dots) \end{aligned}$$

or

$$\begin{aligned} & \rho_o + \rho_{int}(1-\rho_o)(1-\rho_{int})\tau^2(1+\rho_{int}^2\tau^2+\rho_{int}^4\tau^4+\dots) \\ & + \rho_x(1-\rho_o)^2(1-\rho_{int})^2\tau^2(1+2\rho_{int}^2\tau^2+3\rho_{int}^4\tau^4+\dots) \\ & - \rho_x(1-\rho_o)^2(1-\rho_{int})^2\tau^2 \end{aligned} \quad (1)$$

Introducing the following relationships,

$$\begin{aligned} (1+\rho_{int}^2\tau^2+\rho_{int}^4\tau^4+\dots) &= \frac{1}{1-\rho_{int}^2\tau^2} \\ (1+2\rho_{int}^2\tau^2+3\rho_{int}^4\tau^4+\dots) &= \frac{1}{(1-\rho_{int}^2\tau^2)^2} \end{aligned}$$

and regrouping terms gives the following form for equation (1):

$$\begin{aligned} & \rho_o + \rho_{int}(1-\rho_o)(1-\rho_{int})\tau^2\left(\frac{1}{1-\rho_{int}^2\tau^2}\right) \\ & + \rho_x(1-\rho_o)^2(1-\rho_{int})^2\tau^2\left(\frac{1}{(1-\rho_{int}^2\tau^2)^2}\right) \\ & - \rho_x(1-\rho_o)^2(1-\rho_{int})^2\tau^2 \end{aligned} \quad (2)$$

However, as a result of the argument presented in the KBr Window Effects section of this paper, the external and first reflections represented respectively by

$$\rho_o + \rho_{int}(1-\rho_o)(1-\rho_{int})\tau^2$$

do not enter the monochromator and so do not enter the signal processing subsystem. Subtracting these terms from equation (2) yields

$$\begin{aligned} & \rho_{int}(1-\rho_o)(1-\rho_{int})^2\left(\frac{1}{1-\rho_{int}^2\tau^2} - \tau^2\right) \\ & + \rho_x(1-\rho_o)^2(1-\rho_{int})^2\tau^2\left(\frac{1}{(1-\rho_{int}^2\tau^2)^2}\right) \\ & - \rho_x(1-\rho_o)^2(1-\rho_{int})^2\tau^2 \end{aligned} \quad (3)$$

which is the total possible error, E, introduced by the KBr window. Evaluating ρ_o in the same manner as in the KBr Window Effects section using Fresnel's formulae gives

$$\rho_o = 0.043$$

It is seen that ρ_o is equal to ρ_{int} found previously at this small incidence angle of $\theta_i = 15^\circ$. Evaluating equation (3), the total possible error due to the KBr window is found by setting

$$\tau = 1 - \rho_{int}$$

$$\rho_x = 1$$

The final quantity is then

$$E = 0.00136$$

which is clearly less than 1% and negligibly small.

APPENDIX B -- Vendors

Listed below are the vendors from which major system elements were purchased.

<u>Items</u>	<u>Vendors</u>
Monochromator; Gratings	GCA/McPherson Instrument 530 Main Street Acton, Mass. 01720
Sodium Ampules	Ventron Company Alfa Division 152 Andover Street Danvers, Mass. 01923
Houston Instrument Superscribe Strip Chart Recorder	Data Devices 1148 Barberry Downers Grove, Ill. 60515
Goniometer; Light Source and Controls; Related Accessories	Harrick Scientific Corp. Croton Dam Road P.O. Box 867 Ossining, New York 10662
MacCleod Gage; Diffusion Pump	Pope Scientific Inc. P.O. Box 495 N90 W14337 Commerce Drive Menomonee Falls, Wi. 53041
Signal Detector	Charles M. Reeder & Co., Inc. 196 Victor Avenue Detroit, Michigan 48203
Collodion for Cleaning Mirrors	Mallinckrodt, Inc. St. Louis, Mo. 63147
Mechanical Vacuum Pump	Sargent-Walsh Scientific Co. 7300 North Linder Avenue Skokie, Ill. 60076
LIAs	EG&G Princeton Applied Res. P.O. Box 2565 Princeton, New Jersey 08540

Items

KBr Windows

Vendors

Wilks Infrared Accessories
140 Water Street
S. Norwalk, Conn. 06854

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